

PATENT

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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CUSTOMER NO. 26645

In re Application of: James D. Burrington et al.

Serial No.: 10/603,644

Examiner: McAvoy, Ellen M.

Filed: June 25, 2003

Group Art Unit: 1797

Title: Gels that Reduce Soot and/or Emissions from Engines

Hon. Commissioner for Patents  
P. O. Box 1450  
Alexandria, Virginia

Sir:

**DECLARATION UNDER 37 C.F.R. §1.132**

I, James Burrington, declare as follows:

The work of the present patent application is my own work and that I am a co-inventor of the patent application 10/603,644 and am familiar with the reference which was used in the rejection thereof.

I am also a co-inventor of the granted US Patent 6,843,916 which discloses lubricant additive gels (column 1, line 44) having an ashless dispersant, an overbased detergent (column 4, lines 20-25 and 29-32) and oil-soluble antioxidants (column 7, lines 22-33).

The subject matter of the granted patent is my own work and I conceived and invented the subject matter in the earlier application.

I further declare that all statements herein made of my own knowledge are true and all statements herein made on information and belief are believed to be true. All data contained herein was collected under my direction and control. I understand that

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**CERTIFICATE OF EFS SUBMISSION (37 C.F.R. § 1.8(a)(1)(1)(C))**

I hereby certify that this correspondence is being filed electronically via the USPTO EFS to the Commissioner for Patents, United States Patent & Trademark Office, in accordance with §1.6(a)(4) on:

5/5/08  
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/Nancy S. Dedek/

Deposited by: Nancy S. Dedek

The gel compositions claimed in the pending application are non-liquid materials which do not flow, generally do not have measurable Kinematic viscosities, and which cannot be pumped, blended or handled as liquid concentrates. The gel compositions of the present invention are significantly different from the sometimes viscous liquids described in US 6,310,010, Higton, et al., (Higton), including those liquids that experience the “large” Weissenberg effect described by the reference. In other words, the “gels” of the present invention are significantly different from the viscous liquids, described at some points as “gels” in Higton, and the data presented in this declaration demonstrates this difference.

Seven examples were prepared and tested for this study. All examples were prepared according to the directions in Higton at Col 17, lines 60-67. Comparative Example 1 and Comparative Example 2 are based on the "Diphenyl Amine" sample and "No Agent" sample in Table I of Higton (see col 18, lines 16-17 of Higton). The reference samples, which are listed as having "large" Weissenberg effects, are repeated as closely as possible using Applicants' additive components where those described in Higton are unspecified or unavailable. Comparative Examples 3, 4 and 5 correspond to Higton-like compositions in which no Weissenberg effect is observed. None of the comparative examples form a gel, as described by the present invention. Inventive Examples 1 and 2 are Applicants' gel compositions that fall within the claims of the present invention. The following table summarizes the formulations of the examples:

[illegible]

Comparative Example 1 was prepared with a 400 TBN calcium sulfonate over-based detergent and a conventional polyisobutenyl succan / polyamine dispersant, combined to have a dispersant:detergent weight ratio of 4:1, a total actives ratio for the dispersant and detergent combined of 42 wt%, and a 5 wt% diphenylamine (DPA) addition based on the weight of the dispersant, all as specified for the "Diphenyl Amine" example in Higton (see col 17, lines 40-55 and col 18, lines 1-20 of Higton).

Comparative Example 2 used the same dispersant and detergent as Comparative Example 1, with the same weight ratio and actives level but with no DPA, as specified for the "No Agent" sample in Higton (see col 17, lines 40-55 and col 18, lines 1-20 of Higton).

Comparative Examples 3, 4 and 5 used the same detergent and a polyisobutylene dispersant identical to that used in Comparative Examples 1 and 2 but with a lower chlorine level. This lower chlorine level dispersant is discussed in more detail below. Comparative Examples 3, 4 and 5 used the same dispersant:detergent weight ratio as used in Comparative Examples 1 and 2, taken directly from Higton, but with actives levels closer to those in the inventive samples.

Inventive Examples 1 and 2 used the same detergent as all the examples above and the low chlorine polyisobutylene dispersant. Inventive Example 1 had a dispersant:detergent weight ratio of 1:1 and Inventive Example 2 had a dispersant:detergent weight ratio of 1:3.

The lower chlorine dispersant was used in the Comparative Examples 3, 4 and 5 and Inventive Examples 1 and 2. The higher chlorine version of the dispersant used in Comparative Examples 1 and 2 is known to enhance viscosity increase and gel formation compared to the lower chlorine versions used in the other examples, including the Inventive Examples. In other words, Comparative Examples 1 and 2 were made with a dispersant known to be more prone to viscosity increase and gel formation while the Inventive Examples were made with a dispersant known to be less prone to such changes. Thus a large increase in viscosity without gel formation provides a more definitive demonstration distinguishing Comparative Examples 1 and 2 from Inventive Examples 1 and 2.

The only other differences between Comparative Examples 1 and 2 and the original examples in Higton are described here. (i) The dispersant used in the Comparative Examples was not the ethylene-butene copolymer based dispersant made by the Koch reaction, as described in the reference but instead a polyisobutylene based dispersant with similar molecular weight and viscometrics, made by succination and subsequent reaction with polyamines. This conventional dispersant was used due to availability and is not believed to have had any significant impact on the results. (ii)

Dil oil was added to Comparative Examples 1 and 2 in order to achieve the percent actives specified for the Higton examples using the above Applicants above-specified ingredients. It is not known if the examples in Higton had exactly the same levels of dil oil present, since Higton specifies level of actives, not dil oil content. This difference is not expected to have had any impact on the conclusions. However, in order to show this difference had no impact on the comparison, Comparative Examples 3, 4 and 5 were prepared with no dil oil, resulting in the different actives levels shown in Table 2.

(iii) The examples in Higton use a 400 TBN magnesium sulfonate detergent while these examples use a 400 TBN calcium sulfonate detergent. The calcium detergent was used due to its availability and has the same TBN and same actives level as the magnesium counterpart. This difference is not expected to have had any impact on the results.

Additional features of the examples are summarized in the following table, showing that the comparative examples conform to the descriptions in Higton:

**Table 2 – Additional Features of the Formulations**

|  | Comp<br>Ex 1 | Comp<br>Ex 2 | Comp<br>Ex 3 | Comp<br>Ex 4 | Comp<br>Ex 5 | Invent<br>Ex 1 | Invent<br>Ex 2 |
|--|--------------|--------------|--------------|--------------|--------------|----------------|----------------|
| Detergent:Dispersant<br>Weight Ratio         | 4:1          | 4:1          | 4:1          | 4:1          | 4:1          | 1:1            | 3:1            |
| Wt% total of the<br>Dispersant & Detergent   | 77.60        | 78.10        | 98.00        | 96.20        | 100.00       | 98.00          | 80.00          |
| Wt% Actives of the<br>Dispersant & Detergent | 41.90        | 42.20        | 54.90        | 55.70        | 54.00        | 54.40          | 45.40          |
| Wt% of DPA with<br>respect to Dispersant     | 5.00         | 0.00         | 0.00         | 4.90         | 0.00         | 0.00           | 0.00           |
| Physical State                               | Liquid       | Liquid       | Liquid       | Liquid       | Liquid       | Gel            | Gel            |

Generally speaking, true gel materials, like those described in the present application, have rheological properties, which can be measured by small amplitude oscillatory shear testing, that significantly differ from viscous liquids. This shear testing technique measures the structural character of the gel and produces a term called the storage modulus (which represents storage of elastic energy) and the loss modulus (which represents the viscous dissipation of that energy). The higher the storage modulus, the more elastic energy the material stores, indicating a more gelatinous material, as opposed to a liquid which will store only a small amount of elastic energy by comparison. The ratio of the loss modulus/storage modulus, which is called the loss tangent, or "tan delta," is generally >1 for materials that are more liquid-like and

generally <1 for materials that are more solid-like. Therefore, the lower the tan delta value of a material, the more solid-like, or gel-like, it is.

The rotational viscosity, storage modulus and tan delta of the samples was measured using a rheometer at 100°C where the samples were prepared and loaded into the test device at the time the blends were prepared and tested until the results for the non-gel materials leveled off, after 10 minutes. The storage modulus and tan delta values were calculated from the rotational viscosity data. Kinematic viscosity was measured at 100°C by ASTM procedure D445 after a five hour blending period at 100°C, where the components were mixed according to the procedure as described in Higton (Column 17, lines 60-65). The viscosity, storage modulus, and tan delta results obtained are presented in the table below:

**Table 3 – Viscosity and Rheological Data**

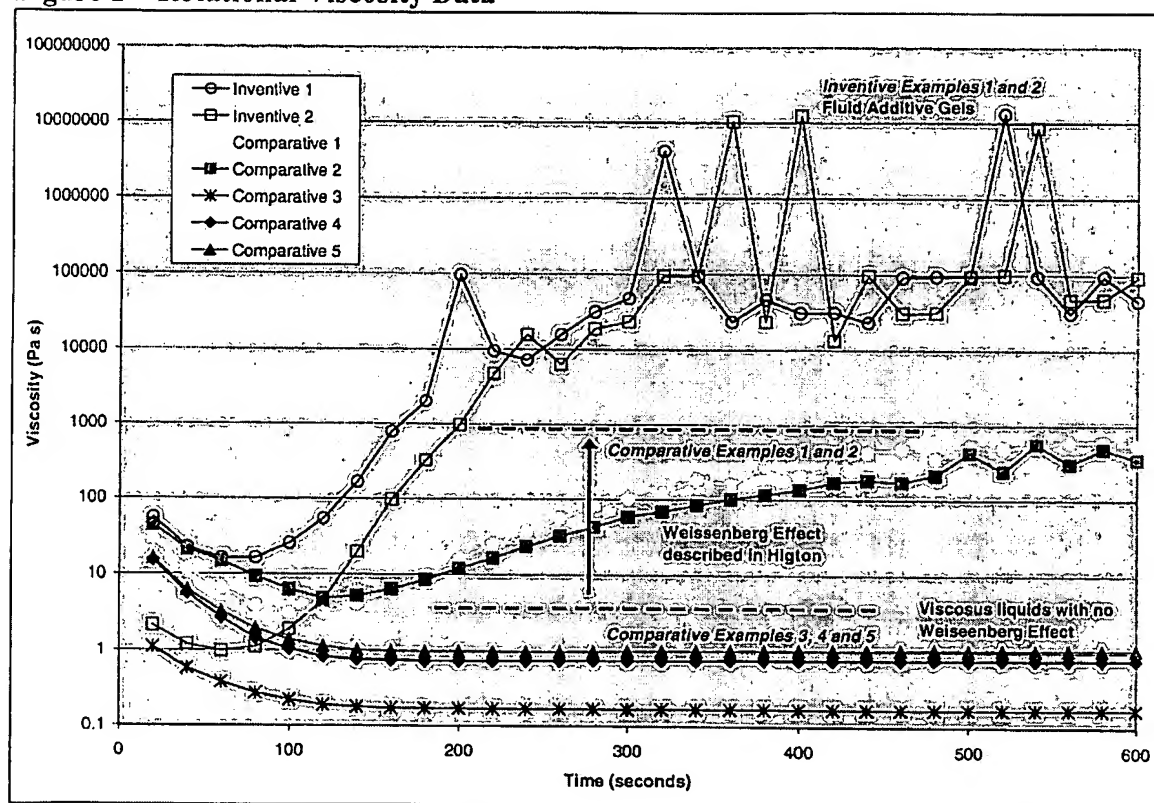
|                        | Comp<br>Ex 1 | Comp<br>Ex 2 | Comp<br>Ex 3 | Comp<br>Ex 4 | Comp<br>Ex 5 | Invent<br>Ex 1 | Invent<br>Ex 2 |
|------------------------|--------------|--------------|--------------|--------------|--------------|----------------|----------------|
| KV at 100C (cSt)       | > 100 k      | > 100 k      | --           | 2611         | 4693         | --             | --             |
| Rotational Visc (Pa s) | 605 k        | 306 k        | 171.7        | 784          | 1063         | >40 MM         | >40 MM         |
| Storage Modulus        | <20          | <20          | <1           | <1           | <1           | >100           | >100           |
| Tan Delta              | 0.94         | 1.11         | >10          | >10          | >10          | 0.22           | 0.15           |

The results show that Comparative Examples 1 and 2 have Kinematic viscosity values similar to those reported for the original examples in Higton. This confirms that the Comparative Examples 1 and 2 are fair representations of the examples in Higton and exhibit at least the same level of viscosities. The results also show that each of the Comparative Examples have rotational viscosities (measured in cPs) consistent with viscous liquids, including Comparative Examples 1 and 2 taken from Higton, which are all significantly lower than the rotational viscosities of the Inventive Examples. The Comparative Examples have rotational viscosities ranging from 170 to 605 k Pa s. The Inventive samples have rotational viscosities above 40 million Pa s, and which also do not level out, as discussed below.

In addition the storage modulus values of the two Inventive Samples are >100 while the values for the Comparative Samples are all <20. The Tan Delta values for the two Inventive Samples are 0.22 and 0.15 while the values for the Comparative Samples are 0.94, 1.11 and higher. All of these properties demonstrate the fundamental differences between the viscous liquids referred to as “gels” in Higton and solid-like fluid additive gels of the present invention.

The differences between the reference materials and the inventive compositions are also shown by the rotational viscosity plots provided in the figure below. The plots are labeled to show the effect of viscous liquids characterized by a rapid increase in viscosity known as the Weissenberg effect. As indicated by the plots, a different effect, which leads to significantly different viscometric properties, takes place in the inventive samples, which form gels.

**Figure 1 – Rotational Viscosity Data**

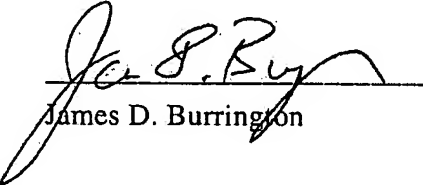


The figure shows that the Inventive Samples have significantly higher rotational viscosity values that reach up well above 10,000 Pa s and continues to bounce with sharp spikes that do not level out over time. This behavior is indicative of gel formation as it shows the material has converted to a elastic, solid-like material. In contrast, the viscous liquids of Comparative Examples 3, 4 and 5 show very smooth rotational viscosity profiles that level off very quickly and which remain below 10 Pa s. As Higton does disclose, some liquids do experience the Weissenberg Effect, which results in a sometimes significant increase in viscosity. This effect is show in Figure 1 in Comparative Examples 1 and 2, which are the worse case scenarios provided in Higton. Here the materials do experience an increase in viscosity which rise to over 100 Pa s but

which then level off and do not rise over 1000 Pa s. There is also none of the sharp spikes seen in the Inventive examples' profiles that are associated with gels. These differences in rotational viscosity profiles clearly demonstrates the fundamental differences between the viscous liquids referred to as "gels" in Higton and the solid-like fluid additive gels of the present invention.

The data presented above shows that there are significant differences between the gel compositions of the present invention and the sometimes viscous liquids described in Higton. Tan delta values are one reliable means of demonstrating this difference, as are the other measurements provided above. The data also shows that the viscous liquids of Higton, including those that experience the Weissenberg Effect and which are sometime referred to as "gels" in Higton, are not true gels, such as those described in the pending patent application.

2 May 2008 (date)

  
James D. Burrington